Modulational Instability in Systems with Integrating Nonlinearity

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(Received 26 October 2004; published 12 August 2005)

A new type of modulational instability for coherent as well as partially coherent light in systems with integrating nonlinearity caused by an irreversible process is investigated both experimentally and theoretically. In such systems plane waves never reach the stationary limit and exhibit a nontrivial time dependence resulting in new features of the modulational instability. For example, the modulational frequency of the nonexponentially increasing perturbation with maximum gain decreases while the wave is propagating. The threshold for vanishing modulational instability due to a finite degree of spatial coherence depends only on system parameters and not on the light intensity.

DOI: 10.1103/PhysRevLett.95.073901 PACS numbers: 42.65.Sf

Modulational instabilities (MIs) attract great attention in many branches of nonlinear science including chemistry, biology, fluid dynamics, plasma physics, and optics [1]. The existence of MI causes a wealth of nontrivial nonlinear phenomena like solitons or patterns (see, e.g., [1–3]). In nonlinear optics, MIs of coherent as well as partially coherent light in dielectric media with instantaneous and noninstantaneous refractive nonlinearity are well understood. The MI in instantaneous Kerr-like media is characterized by an exponential increase of a modulated perturbation with the maximum instability gain, which prevails over all other perturbations with smaller gain [4]. Finally, a pattern of solitonic filaments emerges. The fixed modulation frequency of the perturbation with maximum gain depends on the light intensity. For increasing intensities larger modulation frequencies are observed. Common spatially extended systems with noninstantaneous nonlinearities exhibit a finite memory, i.e., the system remembers all incoming light during a given time period depending on its relaxation time. For small relaxation times the MI of stationary plane waves in instantaneous media is reobtained. But the features of MI change for large relaxation times. In particular, almost all perturbations with different spatial frequencies have approximately the same maximum instability gain; i.e., there exists no distinct favored spatial frequency for the perturbation [5]. Because the initial perturbation consists of dynamic noise, no growth of an instability was predicted and experimentally verified in this case, although a positive instability gain exists [5]. Furthermore, it was shown that even a negative instability gain for all spatial frequencies may occur, i.e., a damping of all possible perturbations, using plane waves with a small enough spatial coherence length [6,7]. The threshold for vanishing MI depends on the incoming light intensity. For larger intensities a smaller spatial coherence length is needed to prevent MI.

Up to now all investigations about MI are realized in nonlinear systems, where the nonlinearity is caused or at least dominated by reversible processes. Contrarily, in this Letter MI is studied in systems with integrating nonlinearity due to an irreversible process. In order to illustrate the differences, let us assume a nonlinear process where the nonlinear refractive index n is governed by the following rate equation: $\partial_t n = g[n]f[I]/T_{\mathrm{exc}} - n/T_{\mathrm{rel}}$, where g and f are arbitrary functions of n and the light intensity I, respectively. The first term on the right-hand side of the equation describes an intensity induced excitation of n within a typical time T_{exc}. The next term is a linear relaxation process with a relaxation time T_{rel}. If the nonlinear process starts at $t = 0$, the refractive index can be written as $n(t) = \int_0^t d\tau g[n(\tau)]f[I(\tau)]\exp[-(t-\tau)/T_{\mathrm{rel}}]/T_{\mathrm{exc}}$. For finite values of $T_{\mathrm{rel/exc}}$ a noninstantaneous nonlinearity with a finite memory time T_{rel} exists. In the case of a fast process ($T_{\mathrm{exc}} \sim T_{\mathrm{rel}} \to 0$) a common instantaneous nonlinearity arises. But for vanishing relaxation, i.e., infinite long relaxation times $T_{\mathrm{rel}} \to \infty$, the nonlinear refractive index is given by the integral of all events in the past $n(t) = \int_0^t d\tau g[n(\tau)]f[I(\tau)]/T_{\mathrm{exc}}$. We term this type of nonlinearity due to an irreversible process an integrating nonlinearity.

From an intuitive point of view one would expect that the MI of nonlinear systems with integrating nonlinearity should be regarded as the limiting case of noninstantaneous systems with very large relaxation times. Consequently, one would deduce that no growth of instability would occur in such systems, at least for coherent light [5]. But, a simple consideration shows that a straightforward transition from large finite to infinite relaxation time is impossible with respect to MI. In case of finite relaxation times a plane wave may reach its stationary limit and MI of such stationary plane waves is usually studied. Contrarily, a strict stationary plane wave can never be reached in systems with integrating nonlinearity. Therefore, MI of a temporally evolving plane wave or, more important, MI of a nonlinear plane wave with a finite starting time has to be studied.

An UV exposed photosensitive polymer exhibits a typical example for a nonlinear system with integrating nonlinearity. In our experimental setup, the beam of an argon-ion laser (363.8 nm) is focused by a microscope objective on a rotating diffuser introducing phase fluctuations ($\sim$1 MHz), which are much faster than the typical response time ($\sim$1 s) of the polymer. The degree of spatial coherence is adjustable and determined by the spot size. The light transmitting the diffuser is collimated by a second objective, in whose back focal point the polymer sample (Ormocer) is positioned. Adding 1 wt % of the photoinitiator Irgacure 369 provides for high UV photosensitivity. Thus an irreversible polymerization reaction with the corresponding increase of the refractive index starts immediately after starting the exposure. A threshold for a minimum amount of deposited energy necessary to start the nonlinear process could not be found [8]. The sample is exposed in the direction of the layer. Because of the curing of the polymer, the emerging MI patterns are fixed and could be investigated after finishing the exposure process using a light microscope, which operates in phase contrast mode.

Let us start with the theoretical description of MI for coherent light in systems with integrating and saturable refractive nonlinearity, such as photosensitive polymers. The propagation of the electric field U is governed by the paraxial wave equation:

$$[i\partial_Z + \tfrac{1}{2}(\partial_X^2 + \partial_Y^2) + N(X, Y, Z, T)]U(X, Y, Z, T) = 0, \tag{1}$$

where the field U is normalized to the field amplitude of the plane wave $|U_0|$. $N(X, Y, Z, T)$ describes the nonlinear refractive index change due to polymerization. It is scaled to the maximum possible index change Δn_s. The transverse and longitudinal normalization lengths read as $x_0 = \frac{\lambda}{2\pi\sqrt{\Delta n_s n_l}}$ and $z_0 = \frac{\lambda}{2\pi\Delta n_s}$, where λ and n_l are the wavelength and the linear refractive index, respectively. For the sake of simplicity the dynamics of the refractive index change is modeled by means of a rate equation describing a one-photon polymerization process empirically [9–11]:

$$\partial_T N(X, Y, Z, T) = [1 - N(X, Y, Z, T)]|U(X, Y, Z, T)|^2. \tag{2}$$

The time T is normalized by $T_0 = 1/(A|U_0|^2)$, where A is the polymerization rate [10]. The analysis given in this Letter can be straightforwardly extended to much more complicated material models. Equation (2) reflects two main points. First, the nonlinearity is a saturable one. The maximum normalized refractive index of $N = 1$ corresponds to a complete polymerization. Second, because the right-hand side of Eq. (2) represents always a positive value, there are no relaxation processes and the irreversible polymerization leads to an integrating, focusing nonlinearity. The plane wave solution of the system of Eqs. (1) and (2) is given by $U_{\mathrm{PW}} = \exp(i[1 - \exp(-T)]Z)$ and $N_{\mathrm{PW}} = 1 - \exp(-T)$, whereby the nonlinear process starts at $T = 0$. The continuous change of the refractive index leads to a time dependent propagation constant of the plane wave. In order to study MI, these plane waves are perturbed by small transverse, spatial modulations as $U = U_{\mathrm{PW}}[1 + \delta U(Z, T, k)\exp(ik_x x + ik_y y)]$ and $N = N_{\mathrm{PW}} + \delta N(Z, T, k) \times \exp(ik_x x + ik_y y)$ with $k^2 = k_x^2 + k_y^2$, $|\delta U| \ll 1$, and $\delta N \ll N_{\mathrm{PW}}$. The linearization of Eqs. (1) and (2) with respect to δU and δN can be written in the form of a single equation for $\delta\tilde{N}(Z, T) = \delta N(Z, T)\exp(T)$

$$\partial_T \delta\tilde{N} = C\cos\left(\frac{k^2}{2}Z + \varphi\right) - 2\int_0^Z \delta\tilde{N}\exp(-T)\sin\left(\frac{k^2}{2}(\zeta - Z)\right)d\zeta, \tag{3}$$

where C and φ denote the amplitude and the phase of the initial field perturbation with the modulational frequency k at $Z = 0$. Using the ansatz $\delta N(Z, T, k) = \sum_{q=0}^{\infty} A_q(Z, k) \times D_q(T, k)$, Eq. (3) can be solved by means of successive approximation analytically; i.e., the $(q + 1)$ component of the series is determined by solving Eq. (3), where the right-hand side is given by the component (q). The series converges very quickly. The larger Z the more addends are required and higher order series components become progressively dominant. In contrast to usual MI, a separation of the Z and the T variables seems to be impossible. As a consequence, there is a nontrivial connection of the Z and T variables for the maximum growing perturbation similar to convective instabilities. During propagation, the time $T_{\max}$ of maximum growth increases [Fig. 1(a)]. The physical interpretation is straightforward inspecting the recursive formula of the temporal shape of the index perturbation $D_{q+1}(T) = \int_0^T D_q(\tau)d\tau\exp(-T)$, which finally leads to the total index perturbation $\delta N(Z, T, k)$ displayed for a given Z and a typical k value in Fig. 1(b). Because of the integrating nature of the nonlinearity the perturbation has to increase for small times. After reaching a maximum at $T_{\max}$ the perturbation has to converge to zero for large times [$\exp(-T)$ term in recursive formula] because the polymerization due to the plane wave tends to the maximum attainable value $N_{\mathrm{PW}}(T) \to 1$ for $T \to \infty$ and the system approaches more and more a linear response. Furthermore, a successive temporal integration according to the recursion formula shifts progressively the temporal index perturbation towards larger times T for increasing q

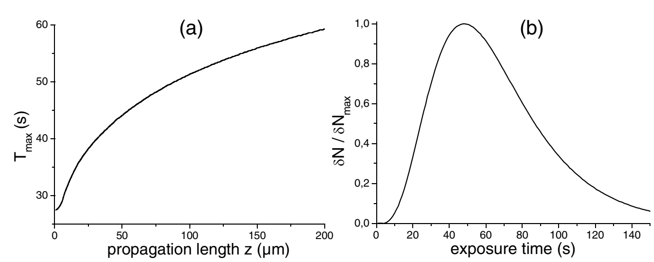

FIG. 1. Evolution of the time $T_{\max}$ of maximum growth (a) of the index perturbation and a temporal shape (b) of such a perturbation ($z = 100\ \mu$m, $k = 0,22\ \mu\mathrm{m}^{-1}$).

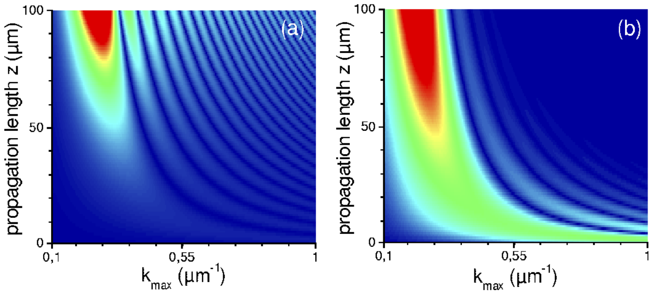

FIG. 2 (color online). Growth of the index perturbation for coherence length $1/\kappa = \infty$ (a) and for $1/\kappa = 1.6\ \mu$m (b).

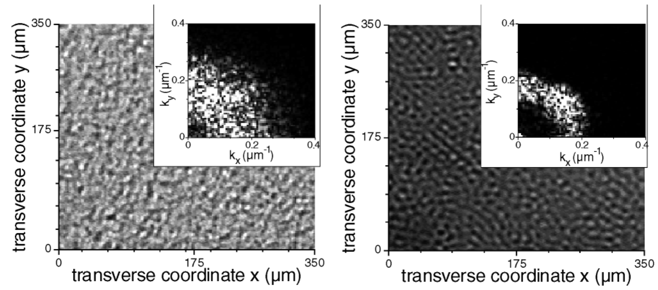

FIG. 3. Transverse MI pattern in Ormocer polymer for coherent (a) and partially coherent (b) exposure, (insets: Fourier analysis).

values. Because higher order series components become progressively dominant for larger propagation distances Z, an increase of $T_{\max}$ occurs during propagation. Thus, the convective character of the MI is an inherent feature of integrating nonlinearity, and it will influence the arising MI pattern considerably as we show below.

In contrast to above expectations [5], the accumulated refractive index perturbation shows a pronounced maximum at a certain modulation frequency $k_{\max}$ for each given propagation length [see Fig. 2(a)]. Moreover, auxiliary maxima occur for higher spatial frequencies. Surprisingly, the value of $k_{\max}$ decreases during propagation [Fig. 2(a)]. This originates from the increase of the values of $T_{\max}(Z)$ with increasing propagation distance as explained above [Fig. 1(a)]. If the maximum growth of the perturbation occurs at larger times, the medium is already polymerized by a larger amount and the potential maximum nonlinear index change for the perturbation decreases. Thus, for increasing values of $T_{\max}(Z)$ the potential nonlinearity for the perturbation decreases continuously. As for all other types of MI, such as in systems with Kerr nonlinearity, a decreasing nonlinearity results in a decrease of the modulational frequency $k_{\max}$ with the maximum growth rate. Figure 3 shows a typical experimental realization of a pattern in the transverse (X, Y) plane, which arises from such a type of MI. It is characterized by a rather broad, ring-shaped spatial spectrum (Fig. 3, inset) as it is predicted by theory. The experimental evidence of the decrease of $k_{\max}$ and a coarsening of the self-organized structure with increasing propagation is given in Fig. 4. Figure 4(a) shows an example of a MI pattern in the (X, Z) plane obtained in Ormocer polymer. Concerning the decrease of $k_{\max}$ during propagation, a comparison of experimental and theoretical results is depicted in Fig. 4(b) for three polymers with different maximum index changes Δn_s. According to the scaling of the transverse coordinates, smaller Δn_s induce smaller spatial frequencies $k_{\max}$.

Now, let us turn towards MI of partially coherent plane waves in systems with integrating nonlinearity. The system is characterized by two different time scales, namely, the coherence time T_c (μs region) due to phase fluctuation caused by the rotating diffuser and the typical response time of the nonlinear medium given by the normalization time T_0 ($10^6\ \mu$s region). Thus, the nonlinearity responds only to the time-averaged intensity and not to the highly speckled light. The resulting partially spatially incoherent light is described by the coherence function $B = \langle U(\vec{R}_1, Z, T)U^*(\vec{R}_2, Z, T)\rangle$ with $\vec{R}_i = (X_i, Y_i)$. The brackets $\langle\,\rangle$ denote the time average over $T_{\rm av}$ with $T_{\rm c} \ll T_{\rm av} \ll T_0$. In this case $\langle NU(R_1)U^*(R_2)\rangle \approx NB$ and $\langle N\rangle \approx N$ holds and the basic equations read as

$$i\partial_Z B(\vec{R},\vec{\rho},Z,T) + \nabla_{\vec{R}}\nabla_{\vec{\rho}} B(\vec{R},\vec{\rho},Z,T) + [N(\vec{R}_1,Z,T) - N(\vec{R}_2,Z,T)]B(\vec{R},\vec{\rho},Z,T) = 0,$$
$$\partial_T N(\vec{R},Z,T) = [1 - N(\vec{R},Z,T)]B(\vec{R},\vec{\rho}=0,Z,T), \qquad (4)$$

where $\vec{R} = (\vec{R}_1 + \vec{R}_2)/2$ denotes the transverse middle point coordinate and $\vec{\rho} = \vec{R}_1 - \vec{R}_2$ the difference coordinate.

The plane wave solution is given by $N_{\rm PW} = 1 - \exp(-T)$ and $B_{\rm PW}(\vec{R},\vec{\rho},Z,T) = b(\vec{\rho})$ with $b(\vec{\rho} = 0) = 1$. For the sake of simplicity the spatial coherence is modeled by an exponential distribution $b(\vec{\rho}) = \exp(-\kappa|\vec{\rho}|)$ according to [6,7]. The correlation length is described by $1/\kappa$. In order to study MI the plane waves are perturbed by small perturbations as $B(\vec{R},\vec{\rho},Z,T) = B_{\rm PW}(\vec{\rho}) + B_1(\vec{R},\vec{\rho},Z,T)$ and $N(\vec{R},Z,T) = N_{\rm PW}(T) + N_1(\vec{R},Z,T)$ with $|B_1| \ll |B_{\rm PW}|$ and $N_1 \ll N_{\rm PW}$. Combining the procedure given in [6,7] and the above

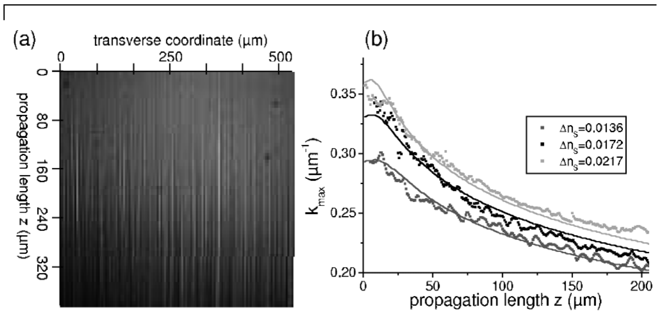

FIG. 4. z-dependent filamentation (a) for coherent exposure and evolution of maximum growing spatial frequency $k_{\max}$ (b) (solid lines: theoretical results).

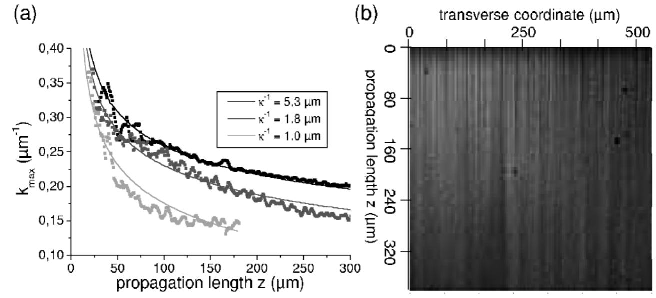

FIG. 5. Evolution of the spatial frequency $k_{\max}$ of maximum growth (a) for z-dependent filamentation (b) for three correlation lengths (solid lines: theoretical results).

described method of successive approximation via series expansion, the resulting linearized equations for B_1 and N_1 are solved analytically. In analogy to the coherent excitation [Fig. 2(a)] a distinct perturbation maximum exists at a modulation frequency $k_{\max}$ for each propagation length [Fig. 2(b)]. But now, those frequencies, which are not favored by the nonlinear system, are damped, resulting in a sharper frequency spectrum and vanishing auxiliary gain maxima, in contrast to coherent excitation. These theoretical considerations are confirmed by the experimental studies. The spatial spectrum of the obtained MI patterns shows a sharp annular structure [Fig. 3(b)]. As above, the favored spatial frequency $k_{\max}$ decreases during propagation. Additionally, $k_{\max}$ critically depends on the spatial coherence length $1/\kappa$ of the plane wave. For a smaller coherence length $1/\kappa$, the stronger diffractive broadening leads to a coarser MI pattern; i.e., smaller values of $k_{\max}$ occur [Fig. 5(a)]. Similar to the investigations in noninstantaneous media with finite relaxation times [6,7], MI can even be eliminated entirely, if the coherence length of the plane wave is smaller than a certain critical value x_c. The theory above predicts a threshold of $1/\kappa = 3.5$ for vanishing MI. In real denormalized units x_c reads as

$$x_c \leq \frac{\lambda_0}{2\pi\sqrt{\Delta n_s n_L}} \times 3.5, \tag{5}$$

where x_c depends only on system parameters and not on the averaged light intensity. In particular, x_c is in inverse proportion to the square root of the maximum nonlinear refractive index change Δn_s. Figure 5(b) shows an experimental realization of a situation just beyond the MI threshold. It can be clearly observed that larger damping rates occur for higher spatial frequencies. Perturbations with small spatial frequencies survive for a rather long propagation distance because the actual coherence length is quite close to the MI threshold.

To conclude, we have shown that a new type of MI exists in systems with integrating nonlinearity. A pronounced maximum of the nonexponentially growing perturbations exists for a certain modulation frequency of the perturbation for each propagation distance leading to spatially structured MI patterns. This is in contrast to the MI in media with a noninstantaneous nonlinearity with finite but rather long relaxation times, where almost all perturbation modes exhibit the same MI gain [5]. Furthermore, the MI shows typical features of a convective instability, i.e., a progressive shift of the maximum perturbation towards larger times during propagation. This leads to a decrease of the spatial frequency with maximum MI gain for increasing propagation length. A decrease of the coherence length results in a decrease of the spatial frequency with maximum MI gain due to stronger diffractive broadening. High spatial frequencies are progressively damped. If the coherence length is smaller than a certain critical value, MI vanishes entirely. This threshold does not depend on the light intensity, but only on system parameters, such as linear and nonlinear refractive indices. All theoretical predictions are verified experimentally in UV exposed photosensitive polymers. Finally, we note that these investigations are of great importance even for the fabrication of homogeneous micro-optical polymer elements [8].

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